Complexity science of multiscale materials via stochastic computations

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SUMMARY

New technological advances today allow for a range of advanced composite materials, including multilayer materials and nanofiber-matrix composites. In this context, the key to developing advanced materials is the understanding of the interplay between the various physical scales present, from the atomic level interactions, to the microstructural composition and the macroscale behavior. Using the developing "Multiresolution Data Sets Mechanics", the "predictive science based governing laws of the materials microstructure evolutions" are derived and melted into a "Stochastic Multiresolution Design Framework."

Under such a framework, the governing laws of the materials microstructure evolution will be essential to assess, across multiple scales, the impact of multiscale material design, geometry design of a structure, and the manufacturing process conditions, by following the cause-effect relationships from structure to property and then to performance.

The future integrated multiscale analysis system will be constructed based on a Probabilistic Complexity Science-Based Mathematical Framework. Its verification, validation and uncertainty quantification are done through carefully designed experiments, and the Life-Cycled Materials Design for products design and manufacturing is performed through the use of Petascale Computing. The various techniques of microstructure reconstruction results in the generation of model microstructures that, at some level, has the same statistical properties as the real heterogeneous media. Having reconstructed the heterogeneous medium, one can then evaluate its effective properties via direct numerical simulation and compare these values to experimentally measured properties of the actual medium. The proposed analysis will be dynamic in nature to capture the multi-stage historical evolvement of material/structure performance over the life-span of a product. In addition to providing more accurate assessment of structure performance with stochastic multiscale analysis, our development will provide the capability of predicting structure failures and system reliability to enable more reliable design and manufacturing decisions in product development.

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1. INTRODUCTION

In the not too distant future, an integrated multiscale analysis system for the design of a reliable engineering structure to sustain harsh environmental conditions within a predetermined lifetime may be possible. In contrast to standard hierarchical homogenization analysis, the multiresolution analysis framework will provide concurrent analysis of critical material, product structure, and
manufacturing decisions in an integrated manner. Such capability is currently missing, but it is critical for achieving integrated material, product, and process design in a multiresolution, multiphysics design environment. The multiresolution mechanics theory is suitable for the analysis of the multiscale/multiphysics material systems such as fuel cells, self healing alloys, high strength alloys, high toughness alloys, microsystems, and thermoelectric materials. The theory connects all the scales dictated by the microstructure in a particular material. These nano/micro scale multifunctional materials are too large for first principles approaches, and too small for conventional FEA. By designing material microstructure intelligently, we may hope to create new materials with desired combinations of strength, toughness and density. The ability to reveal the complexity science of multiscale materials require new methods that can be used to predict macroscale properties accurately based on microstructure (and nanostructure) without resorting to empiricism. A rigorous mathematical framework for multiscale modeling and material optimization will help to make super-lightweight, ultra-strength, low-wear materials a reality of everyday life for energy related, industrial, and medical applications alike. The goal in materials science is to develop a comprehensive understanding of microstructure-properties relationships in order to systematically design materials with specific desired properties. As we envision, a materials scientist starts with a prototype materials sample and uses advanced experimental and imaging techniques to create digital data sets of material microstructure evolutions at multiple scales.

Figure 1. Illustration of increasing image resolution.

Figure 2. Multiresolution and microstructure.

Man made functional materials are modeled by empirical relations rather than fundamental governing laws for their mechanical, electrical and thermal properties. By combining materials science, computer science, image processing, mathematics, and experimental results, we want to derive the governing laws for the materials microstructure evolutions via a multiresolution data set analysis. Experimental techniques such as LEAP, FIB, SEM, and AFM † create digital data

†The list of used abbreviations is located at end of the paper.
sets representative of the microstructure. By way of illustration, images of Fig. 1 are provided by satellites with increasing image resolution. A small number of pixels per km² are found to be sufficient for a global image, whereas the observations of details such as cities, large avenues, buildings and so forth require more and more resolution. In analogous manner, to accurately describe the microstructure of a given material, it is necessary to increase fields and constitutive resolution. Thus, all individual micro-constituents are observable at the very fine resolution (See Fig. 2). These data sets extend down to the quantum mechanical scale and can often be generated from first principles calculations. These data sets of microstructures may be mapped to a continuous resolution axis to make the varying materials length scales more accessible. The microstructure evolution cycle of a component subjected to harsh environments (radiation, thermal-mechanical, etc.) is a stochastic design problem. Modeling materials degradation during these conditions must accounted for stochastic variations in the microstructure that may critically affect life time. The experimental techniques mentioned above provide digital data set from material samples which allow the digital reconstruction of the material microstructure including large particles, small particles and individual micro-constituents (Fig. 3). The achievement of a realistic three dimensional computational model with a highly accurate representation of a random heterogeneous material is a challenging problem. In particular, when using FEA the mesh generation could turn out to be delicate. Indeed, the problems which have to be solved are (i) the preservation of the advantages of a structured mesh without loosing the exactness of the geometric representation, especially when a non-conforming mesh interface is used, and (ii) the mesh distortion due to large displacement motion at particle-matrix interfaces.

In cooperation with QuesTek, using FIB and SEM, as shown in Fig. 4, we can extract a digital data set from the material samples and then reconstruct the material microstructures digitally to include microvoids, inclusions, and defects. The modeling error, that is missing information, will be compensated by uncertainty quantification (section 4). Our vision for deriving the governing laws of life cycle functional materials design is to treat large experimental and computational data sets as the building blocks. Note that the data is in 4-dimensions, \( x, y, z \) and \( t \), so the resolution axis is the 5th dimension and the microstructure is shown in continuous resolution. To represent the stochastic nature of the data, another dimension will be added for each parameter with an axis as the variances of that parameter. The derivation of the mathematical model will involve testing, imaging, characterization, verification and validation, and uncertainty qualification. While materials science and experimental observation will heavily influence the model, it will ultimately be the new predictive science mathematical theory that will unify these ideas and lead to a solution. We envision that the data set mathematical foundation will start with linking spatial scales for continuous resolution of a microstructure. We want to be able to zoom into a microstructure in the same way that modern satellite technology allows us to zoom into images anywhere, anytime, and with any resolution. Hence, the separation of data by scales is done through the use of computer imaging and materials science knowledge, as illustrated in Fig. 4. The quantification and variability of microstructured data sets are performed through the use of statistical and decision making theories, whereas the scale linking is done via testing and characterization of the data set samples.

Finally, to establish the governing laws of these very heterogeneous microstructure evolutions subject to extreme environments, we start with the known single scale homogeneous thermal-mechanical, electrical and mass diffusion laws. We then need to use multi-scale physics to design better multi-functional materials and use the above refined multiresolution data sets to develop a technique to extract the missing information that would otherwise remain hidden in the results of the carefully designed experiments. A good start is to link the missing science with existing single scale governing laws by introducing microstructure fluxes, identifying microstructure transition events, and linking scales by introducing microstructure couple fluxes. In this context, models of random media can be efficiently used to derive an estimation of the macroscopic behavior of a physical system from the knowledge of the microscopic behaviors of its constituents [33, 34, 71]. Models of random media can also be used to obtain predictive tools based on fracture statistics. However, in contrary to the effective properties, damage and fracture phenomena are very sensitive to local defects and the intrinsic scatter in fatigue life in many structural materials is a well known observation [33].

We have demonstrated some successes in achieving the aforementioned objectives through research funded by the NSF/Sandia and the on-going ONR DARPA D3D Digital Structure
programs for the design of high performance steels and titanium alloys. A computational predictive science based multisresolution theory has been developed which overcomes one of the fundamental shortcomings of conventional mechanics of functional materials, which is the inability to predict the microstructure-property relation and its evolutions. The proposed theory can predict the scale and magnitude of inhomogeneous deformation in materials which exhibit deformation at several distinct scales. The theory will remain simple enough, but the “multiresolution” capability of the theory is achieved by extending stochastic theories to multiple nested scales (or resolutions). The proposed mathematical foundation of multisresolution structure (data sets–properties relations) is required to solve the extended multisresolution governing equations [58, 92, 65, 87, 64]. This has already been used to formulate and validate a multisresolution modeling approach for (i) a voided alloy, (ii) an alloy with two scales of embedded particles, and (iii) cemented carbide (WC-Co) which undergoes three distinct scales of inhomogeneous deformation. The embedded particles alloy toughness design considerations for adiabatic shear bands with the additions of grain scale dislocation hardening and thermal softening (heat conduction) effects are also being investigated. The multiscale analysis originates with quantum mechanical design using a density functional theory for microstructures implemented via the FLAPW code, permitting the calculation of bonding energies and other relevant data.

The paper is organized as follows:

- The next section presents and describes tools for processing of random heterogeneous materials for (i) the prediction of effective properties of materials (change of scale); and (ii) the construction of damage and fracture statistics models. As an example, the so-called windowing approach is applied to raw model 4330 to highlight the dependence of the effective properties, namely the effective macroscopic stress versus macroscopic effective strain, on the size and on the location of the considered window.
- The multi-scale theory proposed and developed by Liu et al. [58, 92, 65, 87, 64] is summarized in section 3. The theory describes the multi-scale character of damage and failure in random heterogeneous materials and gives rise to a set of equations that govern the physics of materials.
failure at each scale of interest.

- In section 4, a systematic procedure to calibrate the multiresolution constitutive model is presented. A Bayesian calibration approach based on statistical information derived from DNS of cell models and limited physical experimental data is proposed. The uncertainty propagation from probabilistic multiresolution constitutive model using stochastic projection is demonstrated.

- Conclusions are given in section 5. Appendices A and B contain some useful tools from mathematical morphology and the presentation of two models for random set, respectively. The definition of the distance function is recalled in appendix C.

**Stochastic multiscale data set**

Separate data by scales with help from material science

Quantification & Variability of data set

Scales linking via testing and characterization of samples

Reconstruction of data set

Refined data set in order of scale: 1, ..., n

Resolution axis

Figure 4. Data Set Mathematical Foundation.

2. PROBABILISTIC MODELS OF RANDOM MEDIA

Due to heterogeneities in materials, two nominally identical samples of a material may exhibit different properties. The reason can often be traced back to how a material was synthesized or subsequently processed. These heterogeneities may be associated with spatial distributions of phases of different compositions and/or crystal structures, grains of different orientations, etc. Materials scientists routinely manipulate these heterogeneities to optimize the properties of materials. The word microstructure refers to the arrangements of the compositional heterogeneities within a material. In this sense, microstructure exists on scales ranging from tens of atoms up to that above which the material behaves as a homogeneous continuum (mm for steel).

The mathematical morphology provides some tools enabling the quantitatively characterization of the geometry of microstructures of heterogeneous materials by extracting different resolutions of information from images, and to get some of their macroscopic properties. In the framework of the mathematical morphology, the recent developments regarding the determination of the the effective properties resort to notions of the variance and integral range. It has turned out that the overall moduli obtained by averaging over small domains of composite material, using a sufficient number of realizations for each of the studied boundary conditions, are not the same, in general, as that obtained by a sufficiently large RVE. Besides, in general, the size of a RVE depends on the investigated morphological or physical property, the contrast in the properties of the constituents, and their volume fraction. In other words, for the same microstructure, the RVE size differs if thermal or elastic properties are considered. It should be kept in mind that contrary to the effective
properties, damage and fracture phenomena are very sensitive to local defects and, for example, the intrinsic scatter in fatigue life in many structural materials is a well known observation. The use of mathematical morphology has produced many applications in several areas of digital image processing, particularly with regards to pattern recognition in binary images. The work on the classification of gray level images using mathematical morphology is still in its early stages. Readers interested by a broad treatment of the mathematical morphology is referred to the pioneer work due to G. Matheron [60] or the most comprehensive and mathematically rigorous reference by J. Serra in [78]. Very useful presentations of the subject may be found in a series of publications by D. Jeulin and co-workers [32, 33, 34] and in [81, 43]. Also a recent review of the problematical of RVE including nonlinear behavior of materials is given by Ostoj-Starzewski [74].

2.1. Brief presentation of stochastic models

A random heterogeneous material is a medium occupying a region of space in \(d\)-dimensional Euclidean space \(\mathbb{R}^d\) \((d = 1, 2, 3)\) which can be specified by a fixed probability space, that is a triplet \((\Omega, \mathcal{A}, \mathcal{P})\) such that any sample of this medium is a realization of a specific random process. \(\Omega\) is the set of all possible realizations (or "outcomes") of the model, \(\mathcal{A}\) is the set of events (an event is a family of realizations), and \(\mathcal{P}\) is a probability measure, i.e. a function defined on \(\mathcal{A}\) that attributes to each event \(\omega \subset \Omega\) in \(\mathcal{A}\) its probability \(p(\omega)\). \(\mathcal{P}\) measures the frequency of occurrence of each event and \(p(\omega)\) is such that \(0 \leq p(\omega) \leq 1\) and \(p(\Omega) = 1\). Each realization \(\omega\) of the random medium occupies some subset \(V\) of \(\mathbb{R}^d\) the volume of which is denoted by \(V\). A physical property such as e.g. porosity, Young’s modulus ... of a random material is a random function \(Z\) that is a family of random variables \(\{Z(x)\}\) with \(x \in \mathbb{R}^d\) or in some subset of \(\mathbb{R}^d\). \(Z\) may depend on time to account for evolving microstructures. For a fixed \(\omega\), the local property \(Z\) (i) may be continuous (porosity, crystals orientation in polycrystal) or (ii) may take on discrete values (fiber composites ...)

A random set is a stochastic model whose realizations are subsets of \(\mathbb{R}^d\). Typical examples of random sets are those obtained by thresholding a random function \(Z\) at a fixed level \(\lambda\)

\[ X = \{x \in \mathbb{R}^d \mid Z(x) \geq \lambda\} \quad (1) \]

Any random set \(X\) can be characterized by its characteristic function \(1_X\), defined as \(1_X(x) = 1\) if \(x \in X\) and \(1_X(x) = 0\) if \(x \notin X\). Specifying the statistical properties of a random set using the spatial distribution of its characteristic function amounts to probing the random set with finite sets of points. Since this idea does not work in all circumstances, Matheron [60] proposed to resort to closed sets (complements of open sets) because random closed sets (RACS) include several classes of models that are very useful for practical applications (e.g. points processes). Simply speaking, following Matheron [60], probing a random set using the avoidance logic leads to the RACS theory, Fig. 5-b), and by doing so a set and its closure are indiscernible.

![Structuring element](image)

Figure 5. Probing a random set \(X\) in view to specify its statistical properties (Lanuéjoule [43])

To characterize a RACS \(X\) we can select a compact set \(K \subset \mathbb{R}^d\) and examine the probabilistic location of \(X\) with respect to \(K\). By way of example, if the subset \(K\) is reduced to the point \(x\), it...
is then possible to know if the point \( x \) is located in the set \( X \) or in its complementary \( X^c \). The same process can be repeated for \( K \) composed of \( n \) points: \( K = \{ x + h_i \} \), where \( (h_i, i = 1, n) \) are \( n \) arbitrary vectors.

Let \( \mathcal{K} \) be the family of the compact subsets of \( \mathbb{R}^d \) and \( \mathcal{K}_X \) the sub-family of compact subsets that hit the set \( X \subset \mathbb{R}^d \); \( \mathcal{K}_X = \{ K \in \mathcal{K} \mid K \cap X \neq \emptyset \} \). \( \mathcal{K} \) can be equipped with the \( \sigma \)-algebra \( \sigma_\mathcal{K} \) spanned by the events \( \mathcal{K}_G \) with \( G \) an open set. Matheron [60] showed that probability measures do exist on \( (\mathcal{K}, \sigma_\mathcal{K}) \). The distribution of the RACS \( X \) is completely specified by such a probability measure.

**Theorem 2.1.1.** The functional capacity \( T \), defined on the family \( \mathcal{K} \), of the RACS \( X \) is

\[
T(K) = P \{ X \cap K \neq \emptyset \} \tag{2}
\]

The functional \( T \) is the RACS equivalent of the distribution function of a random variable. By way of illustration, for the particular case for which the RACS \( X \) is made up of a single point uniformly located within a compact subset \( K_o \), its functional capacity reads

\[
T(K) = \frac{|K_o \cap K|}{|K_o|}, \quad K \in \mathcal{K} \tag{3}
\]

Conversely, a RACS whose functional capacity is specified by (3) consists of a single point uniformly distributed within \( K_o \). In practice, various geometrical figures are used for the compact set \( K \), in order to test various morphological properties of a structure. For a given model, the functional \( T \) is obtained either by theoretical calculation or by estimation on simulations of the random structure (on samples of the actual structures). The estimation is easily performed from the implementation of the basic operations of mathematical morphology, namely erosions or dilations. After validation of the model from available data, it is possible to use a predictive implementation of its properties as for example in stereology where 3D properties are deduced from 2D observations [84].

### 2.2. Covariogram, covariance and integral range

The covariogram of a set is the volume of the intersection of this set and its translated by a given vector. The covariogram is, in general, suitable to study the dispersion. A very useful notion aimed to predict the variability of physical properties of a random material as a function of the geometry of its constituents is the integral range. When the set under study is modeled by a stationary RACS \( X \), the notion that corresponds directly to the covariogram is the covariance which turns out to be nothing else than its probabilistic version.

**Definition 2.2.1.** The covariance function \( C_X(h) \) is the probability for the points \( x \) and \( x + h \) to be in the set \( X \):

\[
C_X(h) = P \{ x \in X, \quad x + h \in X \} \tag{4}
\]

The properties of the covariogram are then enriched by stochastic interpretations [79]. \( C(h) \) is estimated from the volume fraction of the set \( X \cap X_{-h} \). In particular, one has \( \lim_{h \to 0} C_X(h) = P \{ x \in A \} = f_X \). Some properties of the covariance are indicated in Appendix A.

Knowledge of the statistical fluctuations of a given stochastic model are required to ensure for example the correctness of any numerical simulation. In this context, it is possible to define a range, namely the integral range, which gives information on the domain size of the structure for which the measured physical properties in this volume have a good statistical representativeness [60, 33, 43].

Let \( Z = (Z(x), x \in \mathbb{R}^d) \) be a second-order stationary random function representing a random physical property of a phase (Young’s modulus, yield stress . . .). Consider the problem of the estimation of the mean \( m \) of a second-order stationary random function \( Z = (Z(x), x \in \mathbb{R}^d) \) with unknown variance \( \sigma^2 \) and correlation function \( \rho \), from the knowledge of one realization \( z(x), x \in V \) of \( Z \) in the bounded domain \( V \subset \mathbb{R}^d \). The average \( \bar{z} = \bar{z}(V) \) of \( z \) over the domain \( V \) which volume is denoted by \( V \), is a realization of the estimator

\[
\bar{Z}(V) = \frac{1}{V} \int_V Z(x) \, dx \tag{5}
\]
the mathematical expectation of which equals $m^\dagger$. To calculate its variance the correlation function $\rho$ between all pairs of points of the domain $V$ is required, namely

$$D_Z^2(V) = \frac{\sigma^2}{V^2} \int \int_V \rho(x - y) \, dx \, dy$$

(6)

The variance $D_Z^2(V)$ does not always vanish when the volume $V$ becomes infinite and subsequently the concept of ergodicity is needed.

**Definition 2.2.2.** The integral range $A_d$ of the random function $Z$ in the space $\mathbb{R}^d$ is the quantity

$$A_d = \lim_{V \to \infty} \left( \frac{D_Z^2(V)}{\sigma^2} \right) V$$

(7)

$A_d$ is homogeneous as a $d$-volume and defined for all usual covariance functions. A simple physical interpretation proposed by Matheron [61] is repeated in Appendix 5.

### 2.3. Generic representation and simulation of microstructures

A very interesting problem in the image analysis of real random media is the reconstruction problem, i.e. the generation of model microstructures that, at some level, has the same statistical properties as the real heterogeneous media. Having reconstructed the random medium, one can then evaluate its effective properties via DNS and compare the obtained results to experimentally measured properties of the actual medium. As mentioned before, random sets are models to describe random geometric microstructures, as they are encountered in applications in natural or technical sciences. In order to fit a random set model $X$ to a given microstructure, specific assumptions on $X$ have to be made. Currently available computational codes can hardly be used to perform systematic investigations of representative microstructures obtained from real samples. Simulations performed on microstructures satisfying some known statistical properties are usually preferred owing to the fact they do not depend on the specific details of the microstructure and they provide generic results. In their study focusing on porous materials, Bilger et al. [7] have considered random microstructures where no particular clustering effect is enforced and clustered microstructures with either connected or disconnected clusters are deliberately introduced. A detailed exposition of the statistics of random microstructures can be found for example in [77, 82]. Herein, the generated family of random microstructures consist of a homogeneous matrix containing circular particles.

Several schemes can be used to generate random microstructures. Circular particles, defined by the coordinates of their centers and their radii, are implanted according to a point process for the coordinates and to a random scalar process for the radius. Depending on the assumptions made for the implementation of the particles, different types of microstructures can be obtained. The classical Boolean model [43, 91] for which the centers are randomly implanted according to a standard Poisson process is disallowed since, for this model, the particles can overlap. To circumvent this occurrence a minimal distance between particles has to be prescribed. This minimal distance is zero for the hard sphere model and is strictly positive for the cherry pit model where particles can neither overlap nor come into contact [82, 7]. For both random microstructure models, particles are generated one after the other: if the current particle does not satisfy the minimal distance requirement, it is removed and a new one is generated and then kept if the constraint regarding the minimal distance is fulfilled. The particle size is not the same for all particles and a uniform probability law for the particles size distribution has been adopted. The overall volume fraction of particles can be monitored precisely. New particles are added as long as the prescribed volume fraction of particles is not exceeded; as soon as it is, the radius of the last particle is reduced until the prescribed volume fraction is reached. The computational method used here requires the unit cell to be periodic.

The constraint on the minimal distance between particles is to be imposed not only between particles located within the unit cell itself but also to particles located in the $n = 8$ adjacent unit cells. When a particle intersects one of the edges of the unit-cell, it is split into two or four

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$^\dagger$The estimator is said to be unbiased.
Figure 6. Statistical analysis of real microstructure of Mod4330 steel.

a) 3D view of submicron particles distribution.

b) Radii distribution (5045 particles)

c) Distribution of the normalized distance function $d_n$: the fast decline of the curve indicates no clustering. Further evaluation is possible by means of thresholding [7].

d) Covariance along the $x$, $y$ and $z$ directions: the asymptotic value is equal to the square of the volume fraction of the submicron particles.
parts. The parts belonging to the unit cell stay in place whereas the parts falling in the peripheral cells are moved into the unit-cell by periodicity. Although the experimental methods devoted to characterize microstructures have recently made a lot of progress, it is still a challenge to derive 3D microstructures from real samples. Further, the obtainment of general results which would not depend on specific details of a given microstructure requires to carry out numerical simulations on microstructures for which some statistical properties can be prescribed and precisely controlled. For these reasons, one chooses to perform numerical simulations on simulated microstructures. Several schemes can be used to generate random microstructures [82, 33].

In this study, simulated microstructures consist of a homogeneous matrix containing circular particles with the same radius $r_j$, the centers of which are randomly implemented such that their distribution is isotropic within the unit cell. Three main different types of microstructure can be obtained, depending on the chosen restriction on the relative positions of the centers, [7]. For the classical boolean model, no restriction is applied on the latter so that voids can overlap. For the hard sphere model, the spheres are not allowed to overlap but still can come into contact. For the cherry pit model a nonzero minimal distance $d$ between the voids is prescribed, preventing both overlapping and contact. In this study, only the cherry pit model is considered. The spheres of the cherry pit model are randomly generated one after the other. If the new sphere does not fulfill the minimal distance requirement with respect to all previous spheres and their periodic reproductions, it is rejected and replaced by a new one until the constraint is satisfied. Periodicity of the microstructure is enforced by splitting spheres which intersect the edge of the image into 2, 4 or 8 parts. The parts which belong to the peripheral cells are moved into the main unit cell by periodicity. The effective volume fraction of particles can be monitored precisely. To this end, the particle radius is derived from the overall volume fraction, from the prescribed particles number and from the known volume of the RVE. The discrete pixels are associated to the matrix or particles, depending on the distance of their centers with respect to the nearest particles center.

2.4. Computational homogenization method based on FEA

The DNS performed on chosen realizations of random materials and the homogenization techniques can be coupled to carry out multiscale simulations of the behavior of materials within the framework of continuum micromechanics. The microstructure computations result in apparent behavior of the heterogeneous material by solving the boundary value problem for the chosen realizations by numerical methods. Beside the FEM, efficient iterative techniques operating by Fast Fourier Transform on periodic media have been used occasionally [72]. The FEM has been driven by the development of mesh generation programs, which lead to the quasi-automatic discretization of the RVE, and by the possibility of implementing complex constitutive equations for the different phases and interfaces. In most of the applications of the FEM to computational micromechanics, the phase arrangements are discretized using standard continuum elements, the mesh being designed in such a way that element boundaries and interface elements are located at all interfaces between constituents. In addition, the microstructure computations provide the local values of the field variables, and thus can take into account accurately the nucleation and growth of damage and the subsequent localization of the strain during the whole process of deformation. When dealing with damage and fracture of materials, these advantages over the homogenization methods which main goal is to yield effective properties, were hindered to some extent by the power of digital computers, which limited the size of the RVE.

The properties obtained through the use of a RVE of finite size subjected to far-field homogeneous deformation depend on the actual boundary conditions. For instance, it is well known that the elastic moduli derived under imposed displacements are always higher than those obtained under imposed traction. The differences decrease as the size of the RVE increases. As an aside, it should be reminded that the response of the RVE must be independent of the type of boundary conditions [28, 76, 31]. More details and standard results dealing with boundary conditions to be prescribed on volume elements and to the definition of effective and apparent properties can be found in textbooks and references such as [73, 41, 97, 50]. A convenient way to numerically solve the homogenization problem is to use i) periodic boundary conditions applied on a periodic unit cell; the two other commonly used boundary conditions are ii) the homogeneous boundary strain rate condition, and iii) the homogeneous boundary stress condition. Periodic boundary conditions
require periodic spatial distribution of the microstructure, and this enables the approximation of
the heterogeneous material by an indefinite extension of a periodic elementary cell in the three
dimensions of space. This assumption has been widely used in the literature as it requires the
modeling of only the highlighted elementary cell, greatly saving computational cost.

\[ \mathbf{v}(x) = \mathbf{L} \cdot x + \mathbf{v}(x) \] (8)

where \( \mathbf{L} \) is the macro-velocity gradient and \( \mathbf{v} \) a periodic velocity perturbation. It is worthwhile
to mention that both homogeneous and periodic boundary conditions may introduce additional
constraints resulting in "biased" numerical solutions including boundary effects and eventually
unrealistic stiff response. Mesarovic and Padbidri [69] have discussed the minimal boundary
conditions applicable to a RVE of any shape. If the simulation of the RVE involves shear, the
condition of homogeneous boundary strain rate is no more applicable since this loading would
preclude localized fields. The only possibility in this situation is to adopt the periodic boundary
conditions. The unknown periodic perturbation field \( \mathbf{v} \) in Eqn. (8) can be eliminated by making the
difference at two points of the RVE in correspondence by periodicity. In addition, many cleverness
available in literature allow to keep constant specific ratios, such as for example the triaxiality,
along the whole processes of deformation of the RVE.

2.5. Windowing approach simulation of a steel alloy

When real microstructures are investigated, generally the experimental data provide us two
dimensional pictures. If 2D simulations are then performed for comparison, the corresponding
microstructures can be either simulated directly from 2D probabilistic processes or obtained as 2D
cross-sections of three-dimensional microstructures. In this paper, by way of illustration, we start
to carry out simulation by using the later method.

Cowie et al. [14] determined that nucleation of micro-voids in a standard 4340 steel occurred
by debonding at the secondary particle-matrix interface. The size and properties of the chromium
(Cr)-based secondary particles in the standard 4340 steel are similar to those examined in the
modified 4340 steel used in the present study and the same nucleation mechanism is expected.
Micro-void nucleation occurs at both titanium carbide (TiC) and fine manganese sulfide (MnS)
secondary particles in the modified 4340 steel. However, only the titanium carbide (TiC) particles
are modeled here. Hao et al. [25] derive a microscale interfacial potential which accounted for the
adverse effects of defects which inevitably exist over a microscale interface. Interfacial debonding
is modeled through a traction-separation cohesive relation implemented at the particle-matrix
interface.

The boundary of a given window is subject to uniform traction and uniform displacement. Such
approach can provide upper and lower bounds on the effective properties of the heterogeneous
materials on hand provided the windows are sufficiently large. Recently, windowing approaches
were extended into the elastoplastic regime, where the bounding properties of the two types of
boundary conditions can be proven within the context of the deformation theory [71]. A quantitative
comparison might be possible by considering more windows with an increased size and containing
more particles. Indeed, using a series of windows of increasing size a hierarchy of bounds can be
generated for a given microstructure, which allows to establish the dependence of the predicted
overall moduli on the size of the microstructure [27, 97]. The number of used windows might be
enough to yield stable value for the ensemble average of the results and to draw at least qualitative
conclusions from these ensemble averages. The dispersion of the results between the considered
windows could be a means to qualitatively evaluate the representativeness of the estimated results.

A mean slice whose dimensions is \( 36 \mu m \times 24 \mu m \times 1 \mu m \) is extracted from the actual
microstructure representing the 4340 steel and shown in Fig. 6-a. The considered slice encompasses
216 secondary particles with non uniform sizes, the distribution of which on the \((x, y)\)-plane is
random without particular clustering. However, along the \(z\)-direction connected clusters are present
and are disregarded. The 2D realization considered for our plain strain simulations include 77
secondary particles and is shown in Figs. 7 and 10. For stereological reasons [84], the 2D particle
area fraction is set equal to the particle volume fraction of the 3D-microstructure (i.e. 0.07%).
In order to obtain effective properties of the realization on hand in terms of, for example, the
Matrix material Carbide particles

<table>
<thead>
<tr>
<th></th>
<th>Matrix material</th>
<th>Carbide particles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young’s modulus $E$ (GPa)</td>
<td>200</td>
<td>600</td>
</tr>
<tr>
<td>Poisson’s ratio $\nu$</td>
<td>0.28</td>
<td>0.30</td>
</tr>
<tr>
<td>Initial yield stress $\sigma_y$ (GPa)</td>
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<td></td>
</tr>
<tr>
<td>Interfacial strength $\sigma_s$ (GPa)</td>
<td>1.0</td>
<td></td>
</tr>
</tbody>
</table>

Table I. Material properties used for the present simulations.

Figure 7. The 2D considered sample comprises 77 particles. Varying window size at the same location of center.

variations of macroscopic equivalent stress versus macroscopic equivalent strain, simulations of arbitrary chosen 2D plane strain volume element (RVE) are performed using ABAQUS finite element software. The size or the position of the mean points of the simulated RVE are changing. The alloy matrix is modeled using an elastic-linear plastic $J_2$ flow plasticity model. The presence of smaller several precipitate particles within the simulated RVE is accounted for. Their number depends on the location and the size of the considered window (see Figs. 7 and 10). The strongly bonded secondary particles to the surrounding matrix are considered to be perfectly elastic, with an elastic modulus consistent with titanium carbide (TiC) [65]. The secondary particles can debond from the surrounding matrix during deformation, via interfacial cohesive relationships creating microvoids. The material properties used for the simulation are given in Table I. Quasi-static fully periodic displacement boundary conditions are applied to the RVE, such that an average tensile strain is applied. The stress triaxiality is kept constant ($\theta := \frac{\Sigma_{eq}}{\Sigma_{eq}} = 3$) during the whole process of deformation on the cell following the description given in [85]. The estimation of material constants at the macro-scale is based on the average response of the RVE. Fig. 7-a and Figs. 10-a represent the RVEs used for the cell model computations in their initial configurations. The accumulated plastic strain contours are shown in Fig. 7-d and Fig. 10-d (RVEs in their deformed configurations). As expected to some extent, the curves ($\Sigma_{eq}, E_e$) shown in Figs. 7-b and 10-b are not coincident, which means that effective properties depend upon the size and/or the position of the RVE used.
Very fined mesh and conforming around the inclusions.

Figure 8. Typical used mesh for the simulation.

Decohesion matrix-particles.

a) The number of secondary particles is 21.

b) The shown curves correspond to windows embedding 8, 15 and 21 secondary particles.

Figure 9. Equivalent plastic strain contours plot and normalized effective stress versus effective strain.

for the cell model computations.

2.6. Statistical description of the size of the RVE

Monte-Carlo simulations were used by Gusev [23] to generate independent realizations of disordered distributions of spheres in a matrix: a few dozen spheres were necessary to obtain small scatter in the averaged property. In Forest et al. [18], the stress-strain curves of polycrystalline wires in torsion were simulated as a function of the number of grains within the cross-section: about 30 grains in the cross-section were necessary to reach stationary responses. Recently, Kanit et al. [36]
Figure 10. The 2D considered sample comprises 77 particles. The size of the window is constant and its location changes.

Figure 11. Equivalent plastic strain contours plot and normalized effective stress versus effective strain.

show that it is possible to derive estimate of the effective properties of heterogeneous elastic materials by computing and ensemble averaging the apparent ones of a sufficient number of volumes containing a given number of grains. These authors have chosen the Voronoï mosaic to represent the 2D/3D morphology of the considered random heterogeneous materials, namely a two-phase random aggregates. A constraint of periodicity at the boundary of the volume of the Voronoï mosaic is superimposed and the FEA is used for the computations. The corresponding parameters
are the constitutive equations of the constituents and the boundary conditions. The computational cost limits the possible number of grains that can be handled in the simulation of one volume element. In particular, such a limit size may be smaller than needed to be a RVE. The obtained physical properties are then not necessarily the effective properties of the volume on hand but merely apparent ones [31, 27]. This statistical approach and the associated computational strategy are developed by Houdaigui et al. [30] to estimate, with a fixed precision, the mean apparent shear modulus and the corresponding dispersion as a function of the number of grains, and are extended to the case of viscoplastic composite materials by Madi et al. [59]. Based on this methodology, it becomes possible to carry out a systematic study of the variance of the results as a function of the size of the investigated domain \( V \). The size of a RVE for an estimated property \( Z \) can be related directly to a statistical precision of the mean value of the results of different realizations for each domain size. If the domain \( V \) is a RVE for the property \( Z \), then theoretically, the dispersion must vanish. In practice, however, one must determine the size of the RVE for (i) a given physical property; (ii) a given relative precision in the estimation of the effective properties; (iii) and a number of realizations that one is ready to generate.

After Matheron [61], the expression of the variance \( D^2 Z(V) \) is

\[
D^2 Z(V) = D^2 Z \left( \frac{A_d}{V} \right)
\]

where \( D^2 Z \) is the local (or point) variance of \( Z(x) \) and \( A_d \) is the integral range of the random function \( Z(x) \) (\( d = 1, 2, \) or \( 3 \)). The scaling law (9) remains valid for an additive combination of the random variable \( Z \) over the region of interest, provided that \( V > A_d \) and \( A_d \) is finite. In case of infinite integral range the scaling law (9) can be replaced in many situations [42, 43] by

\[
D^2 Z(V) = D^2 Z \left( \frac{A_d}{V} \right)^\alpha
\]

with \( \alpha \neq 1 \), so that

\[
\log(D^2 Z(V)) = -\alpha \log(V) + \left( \log(D^2 Z) + \alpha \log(A_d) \right)
\]

by taking logarithms of both left and right-hand sides of (10). An algorithm to estimate the integral range of a stationary, ergodic random function is proposed in [43] starting from a realization \((z(x), x \in V)\). When the number \( N \) of realizations is sufficiently large the mean apparent property \( \bar{Z} \) lies in the interval of confidence

\[
[Z^e(V) - \frac{2 D_Z(V)}{\sqrt{N}}, Z^e(V) + \frac{2 D_Z(V)}{\sqrt{N}}]
\]

Consequently, the relative precision \( \epsilon_r \) on the mean value \( \bar{Z} \) is

\[
\epsilon_r = \frac{2 D_Z(V)}{\sqrt{N Z^e}} = \frac{2 D_Z \left( \frac{A_d}{V} \right)^\alpha}{\sqrt{N Z^e}}
\]

Provided that the variance function \( D_Z(V) \) is known, based on (13), the size of the RVE can be defined as the volume for which \( N \) realizations are sufficient to estimate the mean property \( \bar{Z} \) with a given relative error \( \epsilon_r \).

3. MULTIRESOLUTION CONSTITUTIVE MODELING OF EVOLVING MICROSTRUCTURE AND MATERIAL INSTABILITY

Traditional continuum mechanics theories have focused on approximating the heterogeneous microstructure as a continuum, which is conducive to a FEM mathematical description. Although this makes large-scale simulation of material much more efficient than modeling the detailed
microstructure, the relationship between microstructure and macroscale properties becomes unclear. For example, in order to monitor the health of an aerospace structure, material models must clearly relate the key underlying microstructural parameters (cause) to macroscale properties (effect). Usual crack growth models do not accurately predict crack initiation or describe crack growth at the sub-microstructure scales. There is no mechanical science-based continuum theory to model various stages of damage evolution (or crack initiation) from sub-micron scale to micron scale to sub-millimeter range. The majority of existing work is focused on the prediction of macrocracks which are detectable using Non-Destructive Evaluation (NDE). Since the damage nucleates and progresses in much smaller length scale than can currently be tracked using conventional NDE tools, the development of a predictive model at those micro, sub-micro scales is of utmost importance. The most severe limitation of the existing modeling system is the inability to seamlessly address the multi-scale nature of the material/structural responses required to predict structural response in transient extreme environments.

As depicted in Fig. 12, the damage evolution at a critical location (hot spot) of a structural system, which is made of two-phase (for strengthened and toughened) alloys or composites, is a combination of microstructural events at multiple scales. In this illustration, the macroscopic state of stress initiates the nucleation and growth of the larger set of microstructures. This microstructural stress field creates an intense strain field between the nucleated voids. In turn, the microstrain field nucleates sub-microvoids growth and coalescence, and the interaction among the multi-scale strain fields will lead to final macroscopic fracture.

Figure 12. Schematic evolution of the damage process in ductile high strength steels showing the interacting strain fields at a material point of the structural system, [88].

The multiresolution continuum mechanics theory proposed and developed in [25, 65, 64, 88] aimed at overcoming the limitations of a conventional continuum approach, higher order/gradient models and direct coupling schemes. The key features of the theory are:
1- it allows macroscale properties (e.g. strength, hardness, and toughness) and structure performance to be predicted directly in terms of the key microstructure parameters including length scales of inhomogeneous deformation.
2- Provides a mathematical link between materials microstructure and structural properties and monitors the health of the structure accounting for the manufacturing tolerances and mission specified operation conditions.

The multiresolution mechanics theory is suitable for the analysis of the multiscale/multiphysics structural systems. These nano/micro scale multi-functional materials are too large for first principles approaches, and too small for conventional FEA. The multiresolution mechanics connects all the scales dictated by the microstructure in a particular material. The macroscale mechanical and physical properties of materials are inherently linked to the underlying microstructure and defects. The multiresolution nature of this theory allows prediction of the evolving magnitude and scale of deformation as a direct function of the changing microstructure and defects. In addition, demand for higher strength and toughness, extended fatigue life and lower weight is constantly increasing. We are particularly interested in establishing a probabilistic design oriented link between the structural integrity and material properties e.g. strength, ductility, fatigue life that are affected by the underlying micromechanics of deformation. A computational predictive science-based multiresolution theory [25, 65, 64] has been developed to overcome one of the fundamental shortcomings of conventional mechanics of functional materials, which is the inability to predict the microstructure-property relation and its evolution. While the standard finite element discretization using hp-finite elements, generalized finite elements, and meshfree particle
methods [57, 56, 47, 48, 6] can be applied within the existing FEM code development, the additional microstructure-property relation can be incorporated through additional “scale” adaptivity.

The proposed multiresolution technique is capable of linking overall material properties to the underlying microstructure via the micromechanics at each scale of interest. The small-scale deformation phenomena which have a profound impact on macroscale properties are captured. The technique is general enough to be used in any material which exhibits different constitutive behavior at each scale. It can be implemented in a general finite element framework. In terms of material design, the multiresolution approach is a method capable of describing the crucial link between material microstructure and properties, and offers the exciting prospect of eliminating the need for expensive and time consuming mechanical testing and prototyping. As industry is pushing for lighter and stronger materials, the multiresolution theory offers an opportunity for optimization of properties in terms of microstructural architecture.

3.1. Review of the multiresolution continuum mechanics theory

The multiresolution theory proposed by McVeigh and Liu et al. [64, 88] is developed in terms of the virtual work rate in a body during deformation. Separate contributions to the virtual internal work rate density are assumed to arise from homogeneous and inhomogeneous deformation. Background information can be found in the references [28, 73, 3]. Continuum mechanics is based on the premise that a heterogeneous body can be approximated as a smooth continuous medium. Mathematically, this is known as homogenization: the variational power density \( \delta p(x) \) at any point in the continuum is approximated as the average power density of a superimposed RVE \( \Omega \) of the actual microstructure. The resulting continuum virtual power density field is written as:

\[
\delta p(x) = \frac{1}{V_o} \int_{V_o} \delta p_m \, dV_o \quad \text{with} \quad \delta p_m = \sigma_m : \delta L_m
\]

where \( V_o \) is the volume of \( \Omega \) at the continuum point \( x \). The internal virtual power can be rewritten in terms of the power conjugate local stress field \( \sigma_m \) and velocity gradient field \( L_m \) within \( \Omega \). The subscript \( m \) refers to local fields which exist within the microstructure \( \Omega \). Using the Hill-Mandel lemma [28]:

\[
\delta p(x) = \sigma(x) : \delta L(x) \quad \text{:=} \quad \delta p^{\text{hom}}(x)
\]

where \( \sigma \) and \( L \) are the average stress and velocity gradient over \( \Omega \):

\[
\sigma(x) = \frac{1}{V_o} \int_{V_o} \sigma_m \, dV_o \quad , \quad L(x) = \frac{1}{V_o} \int_{V_o} L_m \, dV_o ,
\]

and \( p^{\text{hom}} \) is the homogeneous internal power density. The mechanical fields \( \sigma \) and \( L \) describe the mechanical response of the continuum point \( x \). In many materials, deformation may become very inhomogeneous at or below the RVE scale, often resulting in a intense strain localization. The conventional homogenization approach given in equations (15) and (16) is fundamentally incapable of representing deformation at these smaller scales. The Hill-Mandel approach has been extended to account for both homogeneous and inhomogeneous contributions such that \( \delta p \) at a material point \( x \) becomes:

\[
\delta p(x) = \delta p^{\text{hom}}(x) + \delta p^{\text{inh}}(x) \quad \text{where} \quad \delta p^{\text{inh}}(x) = \frac{1}{V_1} \int_{V_1} \delta p_m \, dV_1 - \frac{1}{V_o} \int_{V_o} \delta p_m \, dV_o
\]

The term \( \delta p^{\text{inh}} \) is the inhomogeneous contribution at a continuum point \( x \) and is defined as the difference between the average virtual power density at the inhomogeneous deformation scale \( \Omega^1 \) and the average virtual power density at the RVE scale \( \Omega \). As the volume \( V_1 \) of \( \Omega^1 \) is smaller than \( V_o \) at the same point \( x \), it can be shown:

\[
\delta p^{\text{inh}}(x) = \frac{1}{V_1} \int_{V_1} \left( \sigma_m : \delta L_m - \sigma(x) : \delta L(x) \right) \, dV_1
\]

It is useful to rewrite the inhomogeneous power density directly in terms of the local inhomogeneous velocity gradient \( L_m - L \):

\[
\delta p^{\text{inh}}(x) = \frac{1}{V_1} \int_{V_1} \beta_m : \left( \delta L_m - \delta L(x) \right) \, dV_1
\]
where the proposed power equivalence relationship:

\[ \beta_m : (L_m - L(x)) = \sigma_m : L_m - \sigma(x) : L(x) \]  

(20)

has been used. This introduces and defines a local microstress \( \beta_m \) as a power conjugate of the local inhomogeneous velocity gradient \( (L_m - L) \). To account for the locally varying nature of the inhomogeneous deformation, it is assumed here that the local velocity gradient \( L_m \) varies linearly at scale \( V_1 \):

\[ L_m \approx \frac{1}{V_1} \int_{V_1} L_m \, dV + \left( \frac{1}{V_1} \int_{V_1} \nabla L_m \, dV \right) : y := L(1) + G^1(x) \cdot y \]  

(21)

where \( y \) is the local position with respect to the center of \( \Omega^1 \). The variables \( L^1 \) and \( G^1 \) are continuum measures corresponding to volume averages of the local velocity gradient \( L_m \) and gradient of the local velocity gradient \( \nabla L \) respectively. After some algebra, \( \delta p^{inh}(x) \) can be shown to be:

\[ \delta p^{inh}(x) = \left( \frac{1}{V_1} \int_{V_1} \beta_m \, dV_1 \right) \cdot \left( \delta L^1 - \delta L(x) \right) + \left( \frac{1}{V_1} \int_{V_1} \beta_m \otimes y \, dV_1 \right) : \delta G^1 \]  

(22)

This expression contains volume averages of the local microstress \( \beta_m \) and its first moment \( \beta_m \otimes y \) at the scale of the inhomogeneous deformation, \( \Omega^1 \). This is clearer when the virtual internal work rate density is rewritten as:

\[ \delta p^{inh}(x) = \beta^1(x) \cdot \left( \delta L^1 - \delta L(x) \right) + \bar{\beta}^1(x) \cdot \delta G^1(x) \]  

(23)

where the continuum microstress \( \beta^1 \) and the microstress couple (or higher order microstress) \( \bar{\beta}^1 \) are defined as:

\[ \beta^1(x) = \frac{1}{V_1} \int_{V_1} \beta_m \, dV_1 ; \quad \bar{\beta}^1(x) = \frac{1}{V_1} \int_{V_1} \beta_m \otimes y \, dV_1 \]  

(24)

The total virtual work rate density is the sum of the homogeneous contribution and the inhomogeneous contribution

\[ \delta p(x) = \sigma(x) : \delta L(x) + \beta^1(x) : \left( \delta L^1(x) - \delta L(x) \right) + \bar{\beta}^1(x) : \delta G^1(x) \]  

(25)

Each of the continuum measures \( \sigma, \beta^1, \bar{\beta}^1, \sigma_m, \sigma_m \) and \( \sigma_m, \sigma_m \) which describe the virtual internal power at a continuum point are written either directly or indirectly in terms of volume averages of the known local fields \( \sigma_m, L_m \) within the superimposed RVE.

The extension of the foregoing analysis to scale \( n > 1 \) of inhomogeneous deformation is straightforward. A microstress and microstress couple can be introduced at each scale of
inhomogeneous deformation such that the inhomogeneous contribution to the internal power density is a summation over each scale:

\[
\delta p(x) = \sigma(x) : \delta L(x) + \sum_{n=1}^{N} \left( \beta^n(x) : (\delta L^n(x) - \delta L(x)) + \bar{\beta}^n(x) : \delta G^n(x) \right)
\]

(26)

where the continuum microstress at scale \( n \) is given by:

\[
\beta^n(x) = \frac{1}{V_n} \int_{V_n} \beta_m \, \text{d}V_n \quad ; \quad \bar{\beta}^n(x) = \frac{1}{V_n} \int_{V_n} \beta_m \otimes y \, \text{d}V_n
\]

(27)

with \( V_n = |\Omega^n| \). The generalized stress and deformation measures can be defined as

\[
\Sigma = [\sigma, \beta^1, \beta^1, \ldots, \beta^n, \bar{\beta}^n] \\
\Delta = [L, (L^1 - L), \nabla L^1, \ldots, (L^n - L), \nabla L^n]
\]

(28)

Let \( b \) and \( t \) be the macroscopic body and traction forces respectively. The body couple stresses and double traction double forces which balance the microstresses within \( \Omega \) and on its partial boundary \( \Gamma_{R^n} \) are denoted by \( B^n \) and \( R^n \) respectively. \( B^n \) and \( R^n \) drive the inhomogeneous velocity gradient \( (L^n - L) \). Confining attention to quasistatic case, it is straightforward to show that the governing equations are given by [87, 64, 88]

\[
\nabla \cdot \left( \sigma - \sum_{n=1}^{N} \beta^n \right) + b = 0 \quad \text{in} \ \Omega \\
\nabla \cdot \bar{\beta}^n - \beta^n + B^n = 0 \quad \text{in} \ \Omega \quad n = 1 \ldots, N
\]

(29)

\[
\left( \sigma - \sum_{n=1}^{N} \beta^n \right) \cdot n = t \quad \text{on} \ \Gamma_t \\
R^n = \bar{\beta}^n \cdot n : (nn) \quad \text{on} \ \Gamma_{R^n} \quad n = 1, \ldots, N
\]

The technique for deriving these governing equations from the standpoint of RVE modeling gives rise to a natural and systematic framework for deriving microstress and microstress couple constitutive relationships. The key to this theory is that several length scales, given by the characteristic sizes of the averaging volumes \( V_n \), are embedded directly via the microstress couple \( \beta^n \) averaging operation in equation (25). Although other higher order theories introduce stress couples, the difference here is that a microstress couple \( \bar{\beta}^n \) is averaged directly at each scale of inhomogeneous deformation. This naturally introduces length scales \( \ell^n \) which are equal to the width of the square averaging volumes \( V_n \).

3.2. Constitutive relationships

To solve the multiresolution continuum governing Eqn. (29) constitutive relationships are required. The load carrying capacity of the material on hand is characterized at various scales by a coupled elasto-plastic model. While the macro-scale material response is based on traditional homogenization techniques [73, 41, 97, 50] or hierarchical homogenization techniques [25], the microscopic and submicroscopic constitutive relations, based on an averaging operation, remain empirical but physically motivated. The rate forms of the constitutive relationships read:

\[
\dot{\sigma} = \dot{\sigma}(L), \quad \dot{\beta}^n = \dot{\beta}^n(L^n - L), \quad \dot{\bar{\beta}}^n = \dot{\bar{\beta}}^n(G^n), \quad n = 1, \ldots, N.
\]

(30)

During the void growth process the rotations of the micro-domains \( \Omega^1, \ldots, \Omega^n \) with respect to the macroscopic domain \( \Omega \) are disregarded in such a way that \( (L^1, \ldots, L^n) \) can be replaced by their corresponding rates of micro-deformation \( (D^1, \ldots, D^n) \), respectively. Direct numerical simulation turns out to be crucial for determining the microscale constitutive relations. Each of the constitutive models required can be calibrated from computational models of the microstructure by averaging at various scales \( \Omega^n \) within a RVE. Details for the calibration procedure can be found in [85, 62, 64, 88] and are not reproduced herein.
The macroscale constitutive relationship, $\dot{\sigma} = \dot{\sigma}(L)$, can be based on for example the GTN constitutive model [83] or the Bammann-Chiesa-Johnston (BCJ) [5] model. As regards the microscale constitutive relationships $\dot{\beta}^n = \dot{\beta}^n(L^n - L)$ and $\bar{\beta}^n = \bar{\beta}^n(G^n)$ with $n = 1, \ldots, N$, McVeigh and Liu [64] have adopted constitutive models which follow the behavior of conventional nonlinear stress-strain relations with elastic and a plastic regimes.

### 3.3. The shear problem

Ductile fracture mechanisms under low triaxiality is still an open problem. Experimental evidence shows that failure in shear strongly depends on the population of secondary particles [14]. However, shear failure on the scale of dispersed grain-refining carbides is not well understood. Therefore, numerical simulations are used based on unit cell calculations as a tool to investigate these mechanisms. Vernerey et al. [86] has investigated the effect of particle distribution by simulating the shear deformation of a cluster of nine secondary particles with RVE volume fraction 2% and particle radii in the range 30-50 nm. The obtained result accentuates the role of particle distribution on the failure of high-strength steel considered (MOD 4330 steel). The failure mechanism observed has been interpreted as a result of microvoid coalescence in a plane whose direction coincides with the shearing direction. As an illustrative example, a two dimensional plane strain quasi-static shear problem is used to show up the microstress and microstress couple behavior and to evaluate the performance of the two-scale material with respect to localization in shear. A DNS of a unit cell model with two small particles is performed. To make the simulation computationally affordable, the size of the shear specimen is scaled down to one micron. Yet, due to the small size of secondary particles (100 nanometers), the typical size of an element is 10 nanometers. The computations are also carried out with the Abaqus FE code, using quadrilateral elements. The number of elements depend on the size of the particles and their mutual position as well. After discretizing the cell with finite elements, one prescribes the cell to a state of pure shear strain (but zero net stress perpendicular to the shear plane) using periodic boundary conditions. The material properties (matrix, particles and interfaces) are the same as those used for tensile simulations (Table I). The Fig 14 illustrates the plastic strain contours in the sheared unit cell during deformation, after decohesion. Once initiated, debonding rapidly propagates along the interface, in a manner similar to a fracture process. The averaging equations (24) are used to compute the microstress and microstress couple constitutive relationships (30) with $n = 1$ (scale of inhomogeneous deformation). As the inhomogeneous deformation increases, the alloy matrix hardens and the resistance to inhomogeneous deformation increases. This manifests as a hardening effect in the microstress response $\beta^1 = \beta^1(L^1 - L)$ and $\bar{\beta}^1 = \bar{\beta}^1(G^1)$. The average responses of the cell are displayed in Fig 14. The variations of the stress, microstress and microstress couple with respect to their arguments exhibit a softening behavior due to severe material damage. Although the number of particles is limited to two, the average responses are different and outline the effect of the geometric data of a RVE.

### 4. STOCHASTIC CALIBRATION OF MULTiresOLUTION MATERIAL CONSTITUTIVE MODEL

To solve the multiresolution continuum governing equations, the constitutive relationships need to be first determined at multiple scales. The constitutive model should be selected considering the physics of deformation and is generally problem specific. For any chosen model, uncertain parameters of the model should be calibrated by combining the data from both the DNS of RVE and the limited physical experiments. One important issue in the calibration of multiscale material constitutive model is how to account for the various kinds of uncertainties existing in the data during the model calibration process, such as the randomness of microstructure of materials and the measurement error. There is a need for developing methods for stochastic calibration instead of the conventional deterministic calibration. The other challenge is the lack of data associated with physical experiments, especially those at finer-scale levels. In this work a Bayesian stochastic model calibration approach is proposed to effectively integrate the limited physical experiment data as well as those generated from the DNS that capture the intrinsic physical relationships of materials.
MUL TISCALE MA TERIALS VIA STOCHASTIC COMPUT A TIONS

Void growth

\[ \phi_p = 1.0 \, \mu m \]
\[ \ell_p = 2.8 \, \mu m \]

Localization of deformation

\[ \phi_p = 0.5 \, \mu m \]
\[ \ell_p = 5.6 \, \mu m \]

\[ \phi_p = 1.0 \, \mu m \]
\[ \ell_p = 5.6 \, \mu m \]

Figure 14. The 2D plane strain periodic unit cell with different sizes and arrangements of two secondary particles. \( \phi_p \) is the particle diameter and \( \ell_p \) is the distance separating their centers.

key material behaviors across different scales.

4.1. Framework of stochastic calibration of multiscale constitutive model

In a generic notation, a material constitutive model can be expressed as,

\[ \sigma^m(t_i) = \sigma^m(\epsilon(t), T(t), \theta), \quad 0 \leq t \leq t_i \]  

(31)
\[ \phi_p = 1.6, 1.4, 1.2 \text{ and } 0.7 \mu m \]

\[ \phi_p = 1.6, 1.4, 0.6 \text{ and } 0.4 \mu m \]

\[ \phi_p = 1.0 \text{ and } 0.5 \mu m \]

Figure 15. The 2D plane strain periodic unit cell with different sizes and arrangements of secondary particles. \( \phi_p \) is the particle diameter.

where \( \sigma^m(t_i) \) denotes the calculated true stress at time \( t = t_i \) by the constitutive model, \( \epsilon(t) \) and \( T(t) \) are the true strain and temperature at time \( t \). \( \theta \) is a vector of unknown parameters in the constitutive model to be calibrated which are a subset of the whole parameter space. In this expression, the history of strain and temperature becomes the controlling (input) variables. It should be noted that rate form can be used instead of Eqn. (31). If we denote the observed true stress in physical experiments as \( \sigma^e \), we can consider the model calibration as a process to find the
Figure 16. Average cell constitutive behavior:
optimal $\theta$ in the following expression:

$$\sigma^n(\epsilon(t), T(t)) = \sigma^{m}(\theta, \epsilon(t), T(t)) + e$$  \hspace{1cm} (32)

where $e$ is the random parameter that captures the measurement error in the experiments. For the purpose of calibration, the material responses are measured at discrete time steps, $i = 1, \ldots, nt$. Usually replicated experiments are desired for the same setting of control parameters, e.g. $(\epsilon(t), T(t))$ to capture the uncertainty in experimental settings, measurement, sample variability and so on. In the lack of physical experiments, data obtained from virtual test using DNS of RVE cell models with simulated uncertainties could be used for calibration. In this case, the results of DNS can be regarded as $\sigma^c$ in Eqn. (32) while the measurement error term $e$ vanishes. In multiresolution continuum theory the constitutive relationship should be obtained in extended scales as explained in Section 3. Those relationships can be expressed as

$$\sigma = \sigma(L(t), T(t), \theta)$$

$$\beta^n = \beta^n(L^n(t) - L(t), \theta^n)$$

$$\tilde{\beta}^n = \tilde{\beta}^n(G^n(t), T(t), \tilde{\theta}^n)$$

where $\theta, \theta^n$ and $\tilde{\theta}^n$ are the multiscale constitutive model parameters which need to be calibrated in a systematic way using DNS of RVE and/or physical experiments. The microstress $\beta^n$ and microstress couple $\tilde{\beta}^n$ together with their work conjugate strain measures $L^n - L$ and $G^n$ can be obtained by multiscale averaging operation in a RVE simulation (Eqn. 21).

To account for the uncertainty propagated from the randomness in microstructural properties and morphology discussed in Section 2, a stochastic calibration framework is proposed in this Section. Standard stochastic calibration approaches include the Bayesian calibration procedure [39] and the maximum likelihood estimation (a frequentist approach) [93]. With stochastic calibration, the calibration parameters are treated as random variables whose distribution and distribution parameters are determined through the calibration process. In this paper, we propose a Bayesian calibration scheme by integrating data from both the DNS and physical experiments.

The DNS models are generated from the information of statistical properties of the microstructure discussed in Section 2. The constitutive relationships in multiple scales are computed from the results of DNS simulations. These relationships are fitted to pre-selected constitutive models by identifying optimal constitutive model parameters in least square sense. From the statistical analysis on the samples of those constitutive model parameters obtained from DNS of RVE samples, we can get the distribution and correlation of those parameters, which serve as the prior of Bayesian calibration. The physical experimental data is used further to get the posterior estimation of the calibration parameters with Bayesian update strategy. This approach can be effectively applied when only limited experimental data are available while a number of DNS are affordable, (Fig. 17).

4.2. Stochastic calibration of constitutive model from DNS of RVE

4.2.1. Computational procedure The procedure to calibrate multiresolution constitutive model using DNS of cell model is described in [64] for one realization of material microstructure. In this work, to account for the randomness in the microstructure, samples of RVE cell model are generated through the procedure described in Section 2 and for each sample of RVE cell model, parameters of constitutive models are identified by fitting the selected constitutive model with calculated relationship. This procedure is specifically denoted as curve-fitting to differentiate from calibration using physical data. Statistical analysis is performed on the ensemble of constitutive model parameters obtained by curve-fitting. The detailed procedure is described as follows:

Step 1: A sample of RVE is generated as explained in Section 2 and loaded under a range of the boundary conditions.

Step 2: The average stress $\sigma$ and strain measures $L$ associated with the deforming RVE are recorded and used to curve-fit a macroscopic constitutive model.

---

Optimal $\theta$ in the following expression:

$$\sigma^n(\epsilon(t), T(t)) = \sigma^{m}(\theta, \epsilon(t), T(t)) + e$$  \hspace{1cm} (32)

where $e$ is the random parameter that captures the measurement error in the experiments. For the purpose of calibration, the material responses are measured at discrete time steps, $i = 1, \ldots, nt$. Usually replicated experiments are desired for the same setting of control parameters, e.g. $(\epsilon(t), T(t))$ to capture the uncertainty in experimental settings, measurement, sample variability and so on. In the lack of physical experiments, data obtained from virtual test using DNS of RVE cell models with simulated uncertainties could be used for calibration. In this case, the results of DNS can be regarded as $\sigma^c$ in Eqn. (32) while the measurement error term $e$ vanishes. In multiresolution continuum theory the constitutive relationship should be obtained in extended scales as explained in Section 3. Those relationships can be expressed as

$$\sigma = \sigma(L(t), T(t), \theta)$$

$$\beta^n = \beta^n(L^n(t) - L(t), \theta^n)$$

$$\tilde{\beta}^n = \tilde{\beta}^n(G^n(t), T(t), \tilde{\theta}^n)$$

where $\theta, \theta^n$ and $\tilde{\theta}^n$ are the multiscale constitutive model parameters which need to be calibrated in a systematic way using DNS of RVE and/or physical experiments. The microstress $\beta^n$ and microstress couple $\tilde{\beta}^n$ together with their work conjugate strain measures $L^n - L$ and $G^n$ can be obtained by multiscale averaging operation in a RVE simulation (Eqn. 21).

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Step 1: A sample of RVE is generated as explained in Section 2 and loaded under a range of the boundary conditions.

Step 2: The average stress $\sigma$ and strain measures $L$ associated with the deforming RVE are recorded and used to curve-fit a macroscopic constitutive model.
Figure 17. Procedure of Bayesian calibration of constitutive parameters using both DNS and physical experimental data.

**Step 3:** The RVE model is examined to identify regions where deformation is strongly inhomogeneous. Inhomogeneous deformation generally arises due to the interactions between microstructural features.

**Step 4:** An appropriate averaging volume is identified at the scale of this inhomogeneous deformation. The averaging volume should capture a linear variation in the inhomogeneous deformation as strain gradients will be computed.

**Step 5:** A relative strain measure \(L^n - L\) is computed as the difference between the average strain in the inhomogeneously deforming region and the average RVE strain computed in step 2.

**Step 6:** The microstress field \(\beta^n\) within the inhomogeneous averaging volume is computed via a power equivalence relationship (Eqn. (20)). This field is averaged over the inhomogeneous averaging volume to find the continuum microstress.

**Step 7:** The relationship between the relative strain and continuum microstress are curve-fitted to the chosen constitutive model. As mentioned at the beginning of this section, the selection of constitutive model is generally problem specific.

**Step 8:** The average strain gradient \(G^n\) is computed within the inhomogeneous averaging volume.

**Step 9:** The average microstress couple \(\bar{\beta}^n\) is computed within the inhomogeneous averaging volume.

**Step 10:** The relationship between \(\bar{\beta}^n\) and \(G^n\) are curve-fitted to an appropriate constitutive model.

**Step 11:** Steps 3–10 are repeated for each clear scale of inhomogeneous deformation within the RVE, \(n = 1, \ldots, N\).

**Step 12:** Steps 1–11 are repeated for each sample of RVE cell model.

**Step 13:** The statistics and distribution of the parameters in the constitutive model are estimated together with the covariance between the parameters.

### 4.2.2. Statistical analysis of parameters in the constitutive model

Let \(\omega_i, i = 1, \ldots, ns\) be the sample configurations of microstructure generated from the statistical information as proposed in section 2. For each \(\omega_i\), the parameters providing the least square fit can be written as

\[
\hat{\theta}^{(i)} = \arg \min_{\theta} \left[ \sum_{j=1}^{nt} \left\{ \sigma_{DNS}(L(t_j), T(t_j); \omega_i) - \sigma(L(t_j), T(t_j); \theta) \right\}^2 \right]
\]

\[
\hat{\theta}^{n^{(i)}} = \arg \min_{\theta^n} \left[ \sum_{j=1}^{nt} \left\{ \beta^n_{DNS}(L^n(t_j), T(t_j); \omega_i) - \beta^n(L^n(t_j), T(t_j); \theta^n) \right\}^2 \right]
\]

\[
\hat{\bar{\theta}}^{n^{(i)}} = \arg \min_{\theta^n} \left[ \sum_{j=1}^{nt} \left\{ \bar{\beta}^{n}_{DNS}(G^n(t_j), T(t_j); \omega_i) - \bar{\beta}^{n}(G^n(t_j), T(t_j); \tilde{\theta}^n) \right\}^2 \right]
\]
Deterministic calibration of multiresolution constitutive model

1. Loading of the RVE $(\beta^0, \beta^n)$

2. Compute generalized stress vector $\Sigma$ and generalized strain rate vector $\Delta$.

3. Calibrate multiresolution constitutive model
   - Macroscale: $\sigma = \sigma(L, \theta, T)$
   - Scale $n$
     $\beta^n = \beta^n((L^n - L), \theta^n, T)$
     $\bar{\beta}^n = \bar{\beta}^n((L^n - L)\nabla, \theta^n, T)$

Step 7 to step 10

Applied to statistical ensemble of cell models

Step 1 to step 12

Figure 18. Stochastic calibration of multiresolution constitutive models using DNS of statistical cell models.

where $\omega_i$ indicates the dependency of stress measures on the specific microstructure configurations in the DNS, $n_t$ is the number of time step. From the ensemble of the above constitutive parameters, $\hat{\theta}(i), \hat{\theta}^n(i), \hat{\theta}^n(i); (i = 1, \cdots, n_s)$, statistics such as moments and probability distribution are estimated. The first four moments of the parameters are calculated as follows

$$
\mu_{\hat{\theta}_j} = \frac{1}{n_s} \sum_{i=1}^{n_s} \hat{\theta}_j(i)
$$

$$
\sigma^2_{\hat{\theta}_j} = \frac{1}{n_s - 1} \sum_{i=1}^{n_s} (\hat{\theta}_j(i) - \mu_{\hat{\theta}_j})^2
$$

$$
b_1\dot{\theta}_j = \frac{1}{n_s \sigma^3_{\hat{\theta}_j}} \sum_{i=1}^{n_s} (\hat{\theta}_j(i) - \mu_{\hat{\theta}_j})^3
$$

$$
b_2\ddot{\theta}_j = \frac{1}{n_s \sigma^4_{\hat{\theta}_j}} \sum_{i=1}^{n_s} (\hat{\theta}_j(i) - \mu_{\hat{\theta}_j})^4
$$

where $\hat{\theta}_j$ denotes the $j$ -th component of macroscale parameter vector $\hat{\theta}$ and $\mu_{\hat{\theta}_j}, \sigma_{\hat{\theta}_j}, b_1\dot{\theta}_j, b_2\ddot{\theta}_j$ are its mean, standard deviation, skewness and kurtosis, respectively. The moments of the $n$ -th scale constitutive parameters can be estimated in the same way. The covariance between
the parameters are estimated as follows:

$$\text{Cov} \{ \hat{\theta}_j, \hat{\theta}_k \} = \frac{1}{ns - 1} \sum_{i=1}^{ns} (\hat{\theta}_j(i) - \mu_{\hat{\theta}_j})(\hat{\theta}_k(i) - \mu_{\hat{\theta}_k})$$

(36)

The covariance between constitutive parameters in different scales can be calculated in similar manner, which results in

$$\text{Cov} \{ \hat{\theta}_j, \hat{\theta}_k^n \} = \frac{1}{ns - 1} \sum_{i=1}^{ns} (\hat{\theta}_j(i) - \mu_{\hat{\theta}_j})(\hat{\theta}_k^n(i) - \mu_{\hat{\theta}_k^n})$$

$$\text{Cov} \{ \hat{\theta}_j^n, \hat{\theta}_k^n \} = \frac{1}{ns - 1} \sum_{i=1}^{ns} (\hat{\theta}_j^n(i) - \mu_{\hat{\theta}_j^n})(\hat{\theta}_k^n(i) - \mu_{\hat{\theta}_k^n})$$

(37)

The error estimates of the moment can be found in literature dealing with errors in Monte Carlo simulation, e.g. [40]. For uncertainty propagation (UP), we need to estimate the (marginal) probability density function (PDF) of the parameters $\theta, \theta^c$ and $\theta^n$ which can be done in several different ways. One way is to use the empirical distribution system such as the Johnson system, Pearson system and so on [35], which fit a PDF with given moments values. Usually up to fourth order moments are used as inputs. Another way is to use the maximum entropy principle (MEP) [19] which provides the least biased estimate possible on the given partial moment information and, in other words, is maximally noncommittal with regard to missing information [13]. The MEP also uses the moments as inputs but theoretically it can accommodate up to arbitrary order of moments. The PDF estimated by empirical distribution systems or MEP can be compared with the histogram of each parameter and can be tested using the $\chi^2$ test or Kolmogorov-Smirnov test [24]. Fig. 19 is an illustration of statistical analysis for constitutive parameters from the ensemble of stress-strain curves obtained from microstructure samples. It should be noted that the example in this illustration deals with only one scale material behavior.

4.3. Bayesian update of constitutive model with limited experiments

The probabilistic constitutive model calibrated through the DNS of cell models are further calibrated with physical experiments. Since in many cases, the data obtained from physical experiments are scarce, in the proposed framework we use the Bayesian calibration which use the statistical information from DNS of cell models as the prior. In a generic notation, the procedure to update model parameters with Bayesian approach can be written as

$$\pi^*(\theta) = \ell(y^e|\theta) \pi(\theta)$$

(38)

where $\pi(\theta)$ and $\pi^*(\theta)$ denote the prior and posterior distribution of $\theta$ respectively, and $\ell(y^e|\theta)$ is the likelihood function for the observed experimental data $y^e$. The calculation of the likelihood function is the most demanding in the parameter update procedure and it is not straightforward in case the model $y^e$ is the material constitutive model. Instead of considering the overall constitutive relationship as our model response by treating some important response, such as material instability points in the constitutive relationship, the difficulty of the calculation can be alleviated. An updated constitutive model based on both numerical simulations and physical experiments can be validated against additional physical experiment data in the prediction domain of interest. To measure the degree of validity, a validation metric which can account for the stochastic nature of the model with limited number of experiments, such as the U-pooling metric [17], can be used.

4.4. Uncertainty propagation from multiresolution constitutive model

The calibrated multiresolution constitutive model presented in Section 3 is used for prediction of material properties and structure performance together with the multiresolution governing equation. The uncertainty of the constitutive model, represented by the PDF and covariance matrix of the calibration parameters, together with uncertainties from other sources, should be appropriately propagated for the prediction of material/structure performance. A variety of
methods can be chosen for UP [44, 21, 52, 53, 55, 24] and in this paper, we propose to use the method of stochastic projection with polynomial chaos expansion. It allows high order representation and the UP with arbitrary probability measure with fast convergence.

4.4.1. Background of polynomial chaos expansion Any square-integrable random quantity $u$ can be represented as closely as possible with a chaos expansion as follows [21]:

$$u(x, t, \omega) = \sum_{i=0}^{\infty} u_i(x, t) \psi_i(\xi(\omega))$$

(39)

where $x, t$ are arguments indicating spatio-temporal dependency, $\omega$ is an element of sample space indicating the involved quantity is random. $\psi$ denotes the expansion basis in terms of independent basic random variables $\xi(\omega)$. In practice, this expansion is truncated at some order $P$ and in many cases used to represent the output responses of stochastic systems while the input uncertainties are more often represented by the Karhunen-Loeve expansion [20]. Once the expansion bases are determined, the procedure of UP degenerates to the procedure of finding the coefficients $u_i(x, t)$. The basis of chaos expansion $\psi_i(\xi(\omega))$ can be built in several different ways. A general mathematical framework to build the bases with arbitrary probability measure was proposed by Soize and Ghanem [80]. The orthogonal polynomial bases have been most widely used since the Hermite
polynomial chaos which corresponds to the Gaussian probability measure was introduced and it has been extended to account for arbitrary probability measure with algorithms of orthogonal polynomials generation, such as the modified Chebyshev algorithm or Stieltjes algorithm [90, 20].

The polynomial chaos expansion (PCE) has been extensively used in the solution of stochastic partial differential equations (PDEs) with intrusive implementation [46, 94] which manipulates the governing equations to transform the original stochastic PDE into coupled deterministic PDEs using the Galerkin method in the random space. Another approach is the non-intrusive approach [29, 2] which treats the models (or governing equations) as black boxes. Although the intrusive application of PCE has been successfully applied to the solution of static problem with elasto-plastic materials by Anders and Hori [4], it is hardly applicable to the solution of the multiresolution continuum governing equation due to the coupled nature of scales and the nonlinearity of the constitutive models. Thus, in this paper, we use the non-intrusive implementation of PCE to quantify the uncertainty of material responses propagated from the microstructure randomness, as described in the following subsection.

4.4.2. Non-intrusive application of PCE with multiresolution continuum theory  As discussed in subsection 4.3, the uncertainties from microstructure are represented by marginal PDFs and the covariance matrix of the multiresolution constitutive model parameters. Let the response of a material be noted by $y$, then it can be written as

$$y = y(\theta, \theta^1, \theta^2, \cdots, \theta^n, \theta^n)$$  \hspace{1cm} (40)

For convenience, let $\mathbf{X}$ denote an extended vector of parameters with size $np$ whose elements are the union of all elements of parameter vectors $(\theta, \cdots, \theta^n, \theta^n)$, with marginal PDFs $f_X(x_i)$, $i = 1, \ldots, np$ and covariance matrix $\text{Cov}(X_i, X_j), i, j = 1, \ldots, np$. Since the input random variables are correlated, Hermite polynomial chaos with Nataf transformation [51, 68] is used to obtain the chaos expansion of $y$. If we do not consider the spatial variability of the parameters, $y$ can be simply expanded by dimensional polynomial chaos as follows:

$$y = \sum_{i=0}^{P} c_i \psi_i(\xi)$$ \hspace{1cm} (41)

In Hermite chaos expansion, the bases $\psi_i$ are the multidimensional Hermite polynomial which can be generated using a generating function [21]. The coefficients $c_i$ can be obtained by stochastic projection utilizing the orthogonality of $\psi_i$ as

$$c_i = \frac{\langle y \psi_i \rangle}{\langle \psi_i^2 \rangle} = \int_{\Omega} y \psi_i f_\xi(\xi) \, d\xi$$ \hspace{1cm} (42)

where $\langle . \rangle$ is the expectation operator and $f_\xi(\xi)$ means the joint PDF of basic random variables, $\xi$ defined in sample space $\Omega$. Since $\xi$ are independent with each other $f_\xi(\xi)$ is simply the product of each component’s marginal PDF. The stochastic projection is a least square estimator of chaos coefficients in terms of $L^2$ norm. The denominator in Eqn. (42) can be analytically calculated and simple close form expression is available for Hermite polynomials. Thus, the primary computational effort resides in evaluating the numerator, which needs to be calculated using numerical integration schemes.

1- Transformation of $X_i$ into independent standard normal variables

The correlated non-normal variables $X_i$ are first transformed into correlated normal variables $Z_i$ with

$$Z_i = \Phi^{-1}[F_{X_i}(x_i)]$$ \hspace{1cm} (43)

where $\Phi$ and $F_{X_i}$ denote the cumulative distribution function (CDF) of standard normal distribution and $X_i$ respectively. The vector $\mathbf{Z}$ has zero mean, unit standard deviation with reduced correlation matrix $\hat{\rho}$ whose entries are the solution of the integral equation

$$\rho_{ij} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left( \frac{x_i - \mu_{x_i}}{\sigma_{x_i}} \right) \left( \frac{x_j - \mu_{x_j}}{\sigma_{x_j}} \right) \phi(z_i, z_j, \hat{\rho}_{ij}) \, dz_i \, dz_j$$ \hspace{1cm} (44)
where $\rho_{ij}$ is the original correlation coefficient which can be calculated from $\text{Cov} \{X_i, X_j\}$ and $\phi$ denotes the bivariate normal density function. Empirical formula to obtain $\hat{\rho}$ and $\rho$ from are available in [51]. Next, the correlated normal variables are transformed into independent normal variable by the Cholesky factorization or spectral decomposition. Thus $z_i$ can be represented as

$$Z = AU$$  \hspace{1cm} (45)$$

where $\hat{p} = [A][A]^T$. From (43) and (45), the mapping from $U$ to $X$ can be written as,

$$x_i = F_{X_i}^{-1}\left(\Phi\left(\sum_{j=1}^{np} a_{ij} u_j\right)\right), \quad i = 1, \ldots, np$$  \hspace{1cm} (46)$$

where $a_{ij}$ is the component $ij$ of matrix $A$.

2- **Sampling in $u$-space and simulation**

To calculate the expectation in the numerator of Eqn. (42), various methods can be utilized, such as sampling based methods such as Latin hyper cube sampling [11] or quadrature/cubature [44]. The comparison of non-intrusive PCE using tensor product quadrature and the Smolyak’s sparse grid [12] based on the Gaussian quadrature [1] are provided in [15]. It is shown that the sparse grid technique is especially useful when the dimensionality of the problem is high. For simplicity of demonstration, the tensor product quadrature is adopted in the following.

The nodes $\ell_i$ and weights $w_i^f$ for standard normal variable, $\xi_i$ in one dimensional Gaussian quadrature can be identified from the Gauss-Hermite quadrature formula [1, 44]. From them, the $np$-dimensional tensor product quadrature formula for arbitrary $f \in C^0$ is defined as

$$Q^n(f) = (U^1 \otimes \cdots \otimes U^{np})(f) = \sum_{j_1=1}^{m_1} \cdots \sum_{j_{np}=1}^{m_{np}} f(\ell_{j_1}^1 \cdots \ell_{j_{np}}^n) \left(w_{j_1}^1 \otimes \cdots \otimes w_{j_{np}}^{np}\right)$$  \hspace{1cm} (47)$$

Clearly, the above product needs $\prod_{i=1}^{np} m_i$, function evaluations. For the calculation of coefficients in Eqn. (42), the evaluation point $(\ell_{j_1}^1 \cdots \ell_{j_{np}}^n)$ in $u$-space is mapped into $x$-space with Eqn. (46) and corresponding constitutive models are determined from Eqn. (33). Using them, the multiresolution continuum simulation can be performed to obtain material response at the quadrature point. Putting $\tilde{y} \psi$ into Eqn. (45), the chaos coefficient $c_k$ can then be calculated as,

$$c_k = \frac{1}{\psi_k} \sum_{j_1=1}^{m_1} \cdots \sum_{j_{np}=1}^{m_{np}} \tilde{y}(\ell_{j_1}^1 \cdots \ell_{j_{np}}^n) \psi_k(\ell_{j_1}^1 \cdots \ell_{j_{np}}^n) \left(w_{j_1}^1 \otimes \cdots \otimes w_{j_{np}}^{np}\right)$$  \hspace{1cm} (48)$$

where $\tilde{y}$ means $y$ transformed into $u$-space.

3- **Calculation of output statistics**

Once all the coefficients in the expansion are known, the statistics of output response can be easily calculated. Using the orthogonality of $\psi_i$ the mean and variance of response variable can be expressed as follows:

$$\langle y \rangle = c_o, \quad \langle y^2 \rangle = \sum_{i_1=1}^{P} c_{i_1}^2 \langle \psi_{i_1}^2 \rangle$$  \hspace{1cm} (49)$$

and the probability distribution can be obtained by performing Monte Carlo simulation on the obtained expansion of Eqn. (41). The stochastic displacement field can be obtained by interpolating PCE of nodal displacement using the finite element shape functions as

$$d(x, \omega) = \sum_{i=1}^{n_{node}} N_i(x) d_i = \sum_{i=1}^{n_{node}} \sum_{i=1}^{P} d_{i,j} N_i(x) \psi_j(\xi(\omega))$$  \hspace{1cm} (50)$$

The stochastic extended velocity gradient fields in MRCT can be expressed in a similar way.

The procedure explained in this subsection is generic enough to be applied to the UP of any black box type functions. However, the number of expansion terms should be carefully chosen considering the affordable accuracy in the calculation of the coefficients. In the derivation of chaos expansion of output response, the spatial variability of constitutive parameters is not considered. This assumption can be justified when the response $y$ is a material response (i.e. physical property) such as fracture toughness and the RVE size is taken close enough to the process zone size. The UP to structural level responses will be presented in a future work.
5. CONCLUSION

The future challenges in the development of efficient methods based on both multiscale analysis and probabilistic approach lead to the capability of predicting the complexity science of structure failures and system reliability. The ability to predict the scale of deformation turns out to be crucial in order to compute the correct macroscale performance. The characterization of random heterogeneous materials including those possessing particle distributions at various distinct scales, the appropriate probabilistic tool is the functional capacity which can be physically interpreted according to its morphological meaning. A wide range of random models is available to simulate the complex morphology of materials at each scale of interest. The experimental approach of random sets, based on measurements obtained, for example, by image analysis or tomography, is fruitful to select appropriate models, to test their validity, and to estimate their parameters. As regard the prediction of effective properties of random materials, based on the notion of integral range, the needed size of a RVE to be used depends on both the desired precision and the number of realizations of microstructure that is reasonable to consider. Moreover, tools from mathematical morphology can also be used to characterize microstructures and to optimize their morphologies with respect to some physical properties (e.g., stiffness and toughness). In addition, the multiresolution continuum theory can predict the evolving scale of deformation due to the changing microstructure without having to perform large scale detailed microstructure level simulations. With this approach, both the hardening regime and softening regime of plastic deformation can be embedded with their corresponding length scales. The plastic strain is thus dispersed in a much more realistic manner, over scales which are related to microstructural features, i.e., the grain size and precipitate particle spacing. To tackle the important complexity science problem of uncertainty propagation across the scales, a general enough procedure based on Bayesian stochastic model calibration approach is proposed. The approach effectively integrates the limited physical experiment data as well as those generated from the DNS.

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REFERENCES

List of principal symbols and abbreviations

Unless otherwise indicated, bold lowercase roman and greek letters stand, respectively, for first-order and second-order tensors, and bold capital letters for fourth-order tensors.

**Symbols**

- \( L \): typical dimension of the structure,
- \( x \): eulerian coordinates of the current point on the macroscopic scale, \( x = (x_1, x_2, x_3) \)
- \( \Omega_x \): RVE centered at point \( x \),
- \( \ell \): size of the RVE \( \Omega_x \)
- \( \ell^1 \): size of the first micro-domain \( \Omega^1 \) (material between primary particles),
- \( \ell^2 \): size of the second micro-domain \( \Omega^2 \)
- \( y^1 \): coordinates of the current point on the sub-microscopic scale, \( y^1 = (y^1_1, y^1_2, y^1_3) \)
- \(|()|\): volume of \( () \), \( V_0 = |\Omega|, V_1 = |\Omega^1| \)
- \( Z \): volume density of a physical quantity,
- \( \bar{Z} \): average at point \( x \) over the domain \( \Omega_x \) of \( Z(y, t) \),
- \( x \): vector of controllable variables (model input variables)
- \( \theta \): vector of unknown calibration parameters
- \( y \): observable quantity
- \( y^e \): experimental observation of \( y \)
- \( T \): temperature
- \( v \): macro-velocity (average velocity over \( \bar{\Omega} \))
- \( \mathbf{L}_m \): velocity gradient field within \( \Omega \)
- \( \mathbf{L} \): macro-velocity gradient (average velocity gradient over \( \Omega \))
- \( \mathbf{L}_m - \mathbf{L} \): local inhomogeneous velocity gradient
- \( \mathbf{L}^i \): micro-velocity gradient
- \( D \): rate of macro-deformation
- \( G^1 \): gradient of the local velocity gradient \( \nabla \mathbf{L}_m \)
- \( \sigma_{\text{ref}} \): local stress field within \( \Omega \)
- \( \mathbf{\sigma} \): average stress over \( \Omega \)
- \( \beta_m \): local micro-stress (power conjugate of \( \mathbf{L}_m - \mathbf{L} \))
- \( \beta^1 \): continuum microstress
- \( \bar{\beta}^1 \): micro-stress couple
- \( \Delta \): generalized vector of strain rate, \( \Delta = [\mathbf{L}, (\mathbf{L}^1 - \mathbf{L}), \mathbf{G}^1] \)
- \( \mathbf{\Sigma} \): generalized stress vector, \( \mathbf{\Sigma} = [\mathbf{\sigma}, \beta^1, \bar{\beta}^1] \)
- \( T \): stress triaxiality ratio, \( T = \frac{\mathbf{\Sigma}_{\text{eq}}}{\mathbf{\Sigma}_{\text{eq}}} \)
- \( \delta P_{(i)} \): Internal power, \( \delta P_{(i)} = \int_\Omega \mathbf{\Sigma} \cdot \delta \Delta dV \)
- \( P \): Probability
- \( X \): RACS
- \( K \): compact closed set (structuring element)
- \( f_X \): volume fraction of RACS \( X \),
- \( 1_X \): characteristic function of RACS \( X \),
- \( T \): functional capacity
- \( K_h \): geometric covariogram
- \( C_X(h) \): covariance
- \( d \): dimension of the space \( \mathbb{R}^d \), \( d = 1, 2 \) or 3
- \( \mathcal{Y} \): domain of the space \( \mathbb{R}^d \)
- \( A_d \): integral range of a random function in the space \( \mathbb{R}^d \)
- \( D^2_\mathcal{Y}(V) \): variance
- \( D^2_\mathcal{Y}(V) \): local (or point) variance of \( Z(\mathbf{x}) \)
- \( \rho \): correlation function between all pairs of points of the domain \( \mathcal{Y} \)
Abbreviations

AFM: Atomic Force Microscope
DNS: Direct Numerical Simulation
FEA: Finite Element Analysis
FIB: Focused Ion Beam
LEAP: Local-Electron Atom-Probe
MEP: Maximum Entropy Principle
MRCT: Multiresolution continuum theory
PCE: Polynomial Chaos Expansion
PDF: Probability Density Function
RACS: Random closed set
RVE: Representative Volume Element
SEM: Scanning Electron Microscope
UP: Uncertainty Propagation

APPENDIX

Appendix A. Some useful tools from mathematical morphology

0 A1. Geometric covariogram

Definition I.0.1. Let $X$ be a body in $\mathbb{R}^d$ with $d \geq 2$ and $X + h$ be its translation by a vector $h$. The geometric covariogram of $X$ is the function

$$K_X(h) := \text{Volume of } \{X \cap (X + h)\}$$

This defines a real valued function on $\mathbb{R}^d$ which is clearly unchanged by a translation or a point-reflection of $X$, i.e. $K_X(h) = K_X(-h)$. It is readily seen that the volume of a random set $X$ is given by

$$|X| = K_X(0) = \sup_{h \in \mathbb{R}^d} \{K_X(h)\} = \left( \int K_X(h) \, dh \right)^{1/2}$$

where the integral is extended over the whole space $\mathbb{R}^d$. The covariogram $K_X$ is a continuous function on $\mathbb{R}^d$ and moreover a definite positive function which compact support is the so-called difference body defined by $X + (-X) := \{x - y \mid x \in X \text{ and } y \in X\}$ which is always inversion symmetric. Thus, for a bounded set $X$, $K_X(h) = 0$ for $h > \Delta$, where $\Delta$ is the diameter of the set $X$. In fact, the entire covariogram of $X$ equals that of $-X$ as well as any translate $X + h$. This means that $K_X$ can determine $X$ at best up to translation and inversion. Using the characteristic function $1_X$ of the set $X$, the covariogram $K_X$ is given by the convolution

$$K_X(h) = 1_X * 1_{-X}(x) = \int 1_X(x) 1_X(x + h) \, dx$$

For example, the curve representing the covariogram of a 3D ball of diameter $a$ is shown in Fig. 20-b). The value of the directional derivative of the function $K_X$ along vector $h$ at the origin ($h = 0$) is nothing but the total variation of $X$ in this direction. Other interesting properties of the geometric covariogram can be found for example in [79, 43].
A2. Covariance

a) The covariance may provide a characteristic length of the microstructure under consideration. Indeed, for \( h = \infty \), the correlation between \( x \) and \( x + h \) vanishes since the two events \( x \in X \) and \( x + h \in X \) are independent and then \( \lim_{h \to \infty} C_X(h) = p^2 \). If the later limit is reached for a finite value of \( h \), say \( h = a_o \), the points with a distance larger than \( a_o \) are not correlated \([60, 33, 78] \). It can then be advanced that \( a_o \) is characteristic of the size of the set \( X \) and is called the range of the covariance.

b) The expression of the distance \( a_o \) is given in term of the covariance by

\[
a_o = \frac{1}{p(1-p)} \int (C_X(h) - p^2) \, dh
\]  

(54)

c) The covariance also provide information about the distribution of the random set \( X \). For example, the maxima of the covariance, when their abcissa are multiple of each other, indicate (pseudo) periodicities of the distribution of the set \( X \). However, it should be mentioned that the covariance is blind to connectivity.

d) The isotropy of a microstructure can be to some extent deduced from the measurement of the covariance along several directions since the covariance depends on the orientation \( \alpha \) in space of the vector \( h \). The microstructure is isotropic if the measured curves are practically coincident.

A3. Simple interpretation of the integral range

Definition 1.0.2. \( Z \) is said to be second-order stationary if (i) each random variable \( Z(x) \) has a finite mean \( \mu \) that is independent of \( x \), and if (ii) the covariance between each pair of variables \( Z(x) \) and \( Z(y) \) is finite and is a function \( C_Z \) which depend only on the vector \( h = x - y \):

\[
\text{Cov} \{Z(x), Z(y)\} = C_Z(x - y) = C_Z(h), \quad x, y \in \mathbb{R}^d
\]  

(55)

The correlation function is nothing but the normalized version of the covariance by \( C_Z(0) \)

\[
\rho(h) = \frac{C_Z(h)}{C_Z(0)}, \quad h \in \mathbb{R}^d
\]  

(56)

Definition 1.0.3. The second-order stationary random function \( Z \) is said to be ergodic if

\[
\lim_{V \to \infty} D_Z^2(V) = 0
\]  

(57)

The above definition states that \( \bar{Z}(V) \) is an asymptotic good estimator of \( m \) in the quadratic mean sense \([43] \). It does not give information about how large the volume \( V \) must be for the variance of \( \bar{Z}(V) \) to be disregarded.

Let \( f_x \) be the volume fraction of a sample \( V \) with volume \( V := |V| \) in an infinite domain \( \left\{ x \in \mathbb{R}^d \mid f_x \geq \frac{|X \cap V|}{|V|} \right\} \) and let \( D_Z^2(V) \) be the variance of the physical property \( Z \). Suppose the volume \( V \) of the sample very large with respect to the integral range \( A_d \). In this case, one has the approximation

\[
D_Z^2(V) \approx \sigma^2 \frac{A_d}{V} \approx \frac{\sigma^2}{N}
\]  

(58)

where \( N \) denote the integer close to the ratio \( \frac{V}{A_d} \). On the other hand, if the quantity \( \frac{\sigma^2}{N} \) is viewed as the variance of an average of \( N \) independent observations with the same variance \( \sigma^2 \), then \( A_d \) may be considered as a reference \( d \)-volume for predicting whether the size of the sample \( V \) is big enough for the estimation of \( \bar{Z} \) to be sufficiently precise. In addition, after G. Matheron, for a microstructure which covariance is \( C_X(h) \), the variance \( D_{f_X}^2(V) \) of the volume fraction \( f_X \) is given by

\[
D_{f_X}^2(V) = \frac{1}{V^2} \iint (C_X(x - y) - f_X^2) \, dx \, dy
\]  

(59)

a) For a large sample, one has \( D_{f_X}^2(V) = f_X (1 - f_X) \frac{A_d}{V} \). The sample is then statistically equivalent to \( x - \frac{V}{A_d} \) uncorrelated ones.

b) The confidence interval of the average of \( f_X \), \( f \pm 2 D_{f_X}(V) \), is a function of \( V \) and \( A_d \). This gives the relative precision \( \epsilon_r \) of the estimation. Or the volume \( V \) to be used to get a given precision can be obtained as a function of \( f_X \) and of \( A_d \).
Appendix B. Two models for random set

B.1 Boolean model [91, 33, 34, 43]
The boolean model is the most famous and most frequently used random set model. It is a mathematically rigorous formulation of the idea of an infinite system of randomly scattered particles. So it is a fundamental model for geometrical probability and stochastic geometry. The basic used assumption is that random sets can be constructed by placing independent random sets at Poisson points and by taking their union. The Boolean model is constructed by means of two components: a system of grains and a system of germs.

(i) The germs are the points \((x_1, x_2, \cdots)\) of a homogeneous Poisson process of intensity \(\theta\).

(ii) The grains form a sequence \(\{\Xi_n\}\) of i.i.d. random compact sets.

Typical examples are spheres, discs, segments, and Poisson polyhedra.

Tessellations [22, 43]
A tessellation is a division of space into small cells which are usually polyhedra in \(\mathbb{R}^3\) (polygons in \(\mathbb{R}^2\)). The two well known tessellations models are the Voronoi mosaic and the Poisson tessellation models. Let us turn to the brief description of the former tessellation. The procedure that is involved in the construction of a Voronoi mosaic can be reduced to the following two steps [22]:

(i) Let \(X\) be a Poisson point process with intensity function \(\theta\) in \(\mathbb{R}^d\). The points of \(X\) are called germs \(g_j\).

(ii) The space is subdivided into cells \(C_i = C_i(g_i, X)\) which are the zones of influence of the germs \(g_i\) of \(X\): \(C_i\) contains all points in space closer to \(g_i\) than to any \(g_j\),

\[
C_i(g_i, X) = \{x \in \mathbb{R}^3 \mid ||x - g_i|| < \min_{h \in X} ||x - h||\} \tag{61}
\]

This builds a Voronoi tessellation of space. The cell model \(C_i\) is a convex polyhedron because it is the intersection of several half-spaces (points closer to \(g_i\) than to \(g_j\) form a half-space).

Appendix C. The distance function

The distance function is one of the statistical tools which allow to study the details of an actual microstructure and in particular to detect the presence of clusters [7].

Definition I.0.4. Let \(x\) be a point in \(\mathbb{R}^d\) and \(X\) be a random set. The distance function from the point \(x\) to the set \(X\) is the Euclidean distance from this point to the nearest point of this set.

\[
d(x) = \min \{d^j \} = \min_{x^j \in X} \{|x - x^j|\} \tag{62}
\]

In practice of random material inspection through image analysis, the point \(x = x_i e_i\) is a generic pixel representing the matrix and \(d(x)\) is the distance from \(x\) to the point located on the boundary of the nearest particle represented by the random set \(X\). If the particles are circular with center \(x^j = x_i^j e_i\) and radius \(r^j\),

\[
d(x) = \min_{(j=1,\cdots,M)} \left( \sqrt{(x_i - x_i^j)^2 + (y_i - y_i^j)^2 - r^j} \right) \tag{63}
\]

where \(M = (8 + 1) \times N\) (periodicity condition). The normalized distance function \(\tilde{d}\) is simply defined by

\[
d_n(x) = \min_{(j=1,\cdots,M)} \left( \frac{d(x)}{r^j} \right) \tag{64}
\]
The distributions of the distance functions $d$ and $d_n$ turn out to be, to some extent, sensitive to the class of microstructure and then can be used as a statistical tool. For example, Bilger et al. [7] have found for the class of microstructures they have considered for their studies, that the width of the distribution of both functions is larger for clustered microstructures than for random ones.